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IRIDESCENT COLOR IN PERISTERITE

BY

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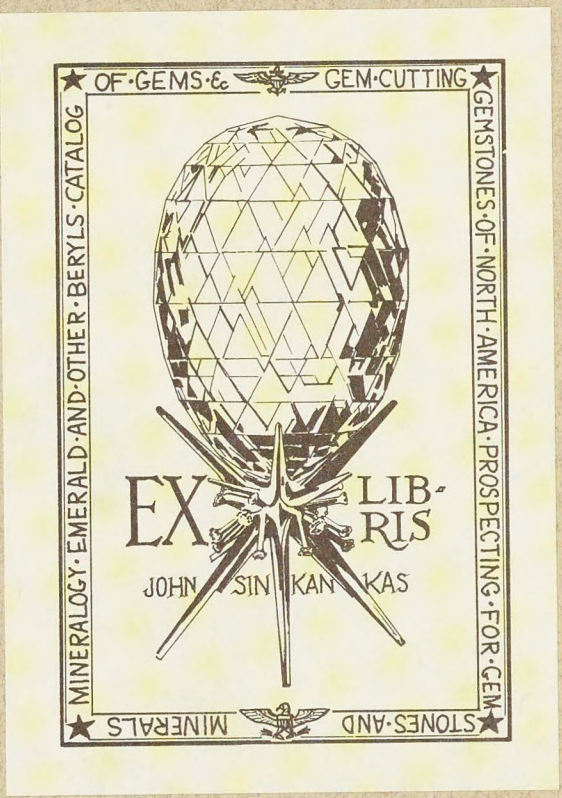
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IRIDESCENT COLOR IN PERISTERITE*

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On this tenth anniversary of the founding of the *Mineralogical Society of America* your president has selected as the topic of his address a field of study in which much fruitless work has been done, and where very little is known. Many terms such as labradorescence, opalescence, iridescence and scattering of light have been applied to the peculiar color phenomenon that is probably best known in labradorite, one of the first minerals to be described from this continent. The last worker in this field after much excellent work frankly acknowledged defeat in trying to reach a satisfactory explanation for the colors. A century and a half has elapsed since labradorite was named and in this period of time no satisfactory explanation of the iridescent color, based on sound physical laws, has appeared.

Inasmuch as the interpretation of the phenomena observed is somewhat radical and may have far-reaching results, the writer has consulted his colleagues in the Department of Physics in the University of Toronto who have assured him that they see nothing in the interpretation that is unsound from a physical point of view.

The subject was selected because during the past summer the writer collected a quantity of peristerite near Hybla, Ontario, which exhibits iridescent color in a remarkable degree. While examining the material it was seen that this color was not visible under ordinary conditions of reflection but that in certain cases the incident light and the reflected iridescent light were both on the same side of the normal to the reflecting surface, while at the same time there was an ordinary reflection with equal angles of incidence

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and reflection. In only two positions was the plane containing the incident white light and the iridescent reflected light perpendicular to the visible reflecting surface. This peculiar optical behavior of the peristerite led the writer to look up the literature bearing on the subject of iridescent colors, more particularly in connection with the feldspars, with the result that he decided to outline our present knowledge of this subject with the addition of certain conclusions of his own.

The same or a similar phenomenon is to be observed in other minerals and artificial crystals, but as it is possible that an explanation that seems good for this mineral may not apply to all, the final discussion is limited to peristerite.

Although certain theories have been advanced to account for these iridescent colors there has been no adequate explanation, nor until quite recently was there any sound basis for reaching a satisfactory conclusion.

It is probably safe to say that no work based on measurements was done on this problem before 1827 when von Hessel did his first work which was mentioned by Nordenskiöld as being the first real effort to solve the problem, so that in general we will not include in our summary of work done the conclusions that were reached without measurements.

Von Hessel[1] who studied this phenomenon in 1827 later made a two circle goniometer and showed not only that the iridescent light was reflected in definite positions but that when the crystal was turned so that the reflecting plane was perpendicular to the polar axis of his instrument the reflection did not disappear in a complete rotation about this axis. He attributed the color to reflections from inner cleavage planes.

Brewster[2] determined that the iridescent color in labradorite was reflected from a plane which made an angle of 10°52′ with the cleavage face *P*. He attributed the colors to reflections from an infinite number of planes and showed that the color of the transmitted light was complementary to that which was reflected.

In showing the path of the light he fails to realize that with his construction there is no possibility of the emerging ray interfering with an incident ray as he assumes normal refraction at the cleavage plane whereas the refraction must be at a plane or

[1] *Kastn. Arch. Nat.*, 1827, **10**, 273; *Pogg. Ann.*, 1850, **79**, 443.

[2] *Trans. Roy. Soc. Edinb.*, **XI**, 322–331, 1829.

pair of planes parallel to the plane of reflection if there is to be interference.

N. Nordenskiöld[3] working with labradorite from Finland determined the position of the plane giving the iridescent reflection and attributed it to cleavages. His material exhibited a zonal arrangement of color which requires further study. The same zonal arrangement was later observed by Schrauff.[4] He also demonstrated that the refractive index was involved in the phenomenon and showed his idea of the path of the light. His most important conclusion was that the atoms of transparent crystals can be

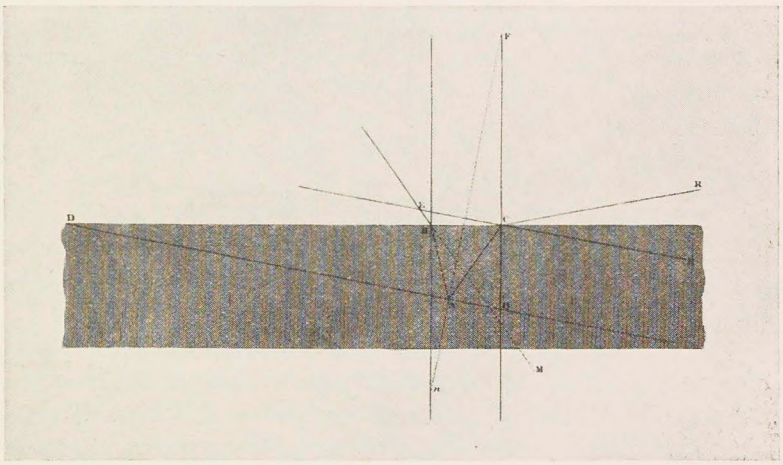

FIG. 1.

arranged for the refraction of light in different ways. With no other foundation and presumably with many doubts and mental reservations, he enunciated the principle that is essential to a logical explanation of iridescent reflections from crystals, but unfortunately in showing the path of the light he used the same general construction as that employed by Brewster.

In the century that has elapsed since Nordenskiöld enunciated this important principle there is no worker in this field who has shown such sound reasoning. It remained for von Laue and a host of workers with X-rays to bring additional information showing

[3] *Kongl. Vet. Acad. Handlinger*, **1829**, 42; *Pogg. Ann.*, 1830, **19**, 179, translated by J. Senff.

[4] *Sitzungsb. Akad. Wiss., Wien*, **60**, 996–1053, 1869.

that reflections can be obtained from a multitude of atomic planes which in general do not coincide with the visible surface.

Senff[5] observed in "Finnish labradorite" that the iridescent color was visible when the crystal was turned $22\frac{1}{2}°$ from the position when the second cleavage, (010), gave its normal reflection.

Bonsdorff[6] attributed the play of colors to very fine and uniform particles of quartz because the labradorite with a play of colors contained more silica than others. This was in the days before the use of the petrographic microscope and was merely *ex cathedra* argument.

Reusch[7] assigned indices to the planes that reflected the iridescent light in labradorite and attributed the reflection to cleavages which were so minute as to be invisible under the microscope. He also said that the color was very probably a low order color from thin plates.

Vogelsang[8] regarded the common blue color of labradorite as a polarization phenomenon due to its lamellar structure, while the golden or reddish schiller, with the other colors, was supposed to be due to the presence of black acicular microlites and yellowish red microscopic lamellae or to the combined effects of these with the blue reflections.

His contention is sound so far as polarization is concerned; for light that has entered the crystal must be doubly refracted and the two rays polarized in planes at right angles to each other. Both rays, however, on reflection emerge from the crystal in the same phase and, in consequence, as was later shown by Stokes, there is no evidence of polarization to be seen with an analyzer.

O. Anderson[9] in his paper on Aventurine Feldspar says:

> "Aventurine feldspars (sunstones) should be sharply distinguished from the other group of color-playing feldspars known as moonstones, murchisonites and labradorites. These feldspars are characterized by a rather subdued, generally bluish or greenish play of colors ("glaukisiren") which is not caused by any visible lamellae but perhaps by submicroscopic inclusions. The colors are probably due to scattering of light by particles

[5] *Pogg. Ann.*, 1829, **17,** 352.
[6] *N. Jahrb.*, **1838,** 681.
[7] *Pogg. Ann.*, 1863, **120,** 95.
[8] *Arch. Néerland.*, **3,** 32, 1868.
[9] *Am. J. Sci.*, (IV), Vol. **40,** p. 351.

smaller than the wave length of light, and cannot be explained as ordinary colors of thin films."

Stokes[10] as the result of the examination of crystals of chlorate of potash enunciated the following points:

(1) If one of the crystalline plates be turned round in its own plane, without alteration of the angle of incidence the peculiar reflection vanished twice in a revolution, *viz.*, when the plane of incidence coincides with the plane of symmetry of the crystal.
(2) As the angle of incidence is increased the reflected light becomes brighter and rises in refrangibility.
(3) The colors are not due to absorption, the transmitted light being strictly complementary to the reflected.
(4) The colored light is not polarized.
(5) The spectrum of the reflected light is frequently found to consist almost entirely of a comparatively narrow band. In many cases the reflection appears to be almost total.

In discussing the cause of the phenomenon he attributed it to twin lamellae of definite thickness.

Rayleigh,[11] in discussing Stokes' work, concluded that the iridescent colors in potassium chlorate crystals came from a large number of reflecting surfaces at regular intervals which he concluded were twin planes. He also showed that in Iceland spar, in which polysynthetic twinning had been artificially induced, iridescent color was to be seen but not when the plane of the incident light coincided with a plane of symmetry of the crystal.

Rayleigh, the younger, shows that the crystals of potassium chlorate were twinned[12] and points out that the colors do not appear at strictly normal incidence.[13] He also shows that mother of pearl consists of alternating layers of calcite and horny matter.[14] In examining labradorite he established a definite plane of reflection for the iridescent light[15] which he distinguished from the reflections from inclusions. He, however, failed to assign indices to the reflecting plane.

[10] *Proc. R. Soc.*, **XXXVIII,** 1884–85, 174–185.
[11] *Phil. Mag.*, Vol. **26,** pp. 256–265, 1888.
[12] *Proc. Roy. Soc.*, Vol. **102,** pp. 668 et seq., 1923.
[13] *Proc. Roy. Soc.*, Vol. **102,** p. 676, 1923.
[14] *Proc. Roy. Soc.*, Vol. **102,** pp. 675 and 677, 1923.
[15] *Proc. Roy. Soc.*, Vol. **103,** p. 37, 1923.

Bøggild, in his treatise on the Labradorization of the Feldspars,[16] states that in the plagioclases this phenomenon is found in only two groups having the composition $92\frac{1}{2}$ to $85\frac{1}{2}$ and 49 to 38 per cent of albite by weight, though the tabulation of his measurements fails to show perfect agreement with his statement. He points out that in the peristerites there is a single plane which gives colored reflections, while with the labradorites there are three planes. Usually only one plane is observed in labradorite but some specimens show two planes which may be any combination of the three possible planes. No case is mentioned where all three planes are shown on a single specimen.

He also observed that the material was either twinned or laminated. To the reflecting plane in albite (peristerite) he indicates positions to which he assigns indices corresponding approximately to $(08\bar{1})$ and $(\bar{1}.21.\bar{2})$. He, however, failed to find a cause for the iridescent color and says:

> "Labradorization is the peculiar reflection of the light from submicroscopical planes orientated in one direction (rarely in two directions); these planes have never such a position that they can be expressed by simple indices, and they are not directly visible under the microscope.
>
> "It must of course be considered perfectly enigmatic what can produce the labradorization in the plagioclases, and equally impossible to explain the peculiar orientation of the lamellae which produce that phenomenon. It is not possible to find any cause for the orientation of the lamellae in these peculiar directions, and there is no other property, neither crystallographical nor optical, which can be connected with them."

An extremely important contribution to our knowledge is, however, given by Bøggild in his concluding paragraph where he points out that gedrite from Avisiarfik in Greenland gives iridescent color from the reflecting plane (010), and that the substance is perfectly homogeneous and there were no reflecting intercalations visible under the microscope. In this case it would appear that twinning does not enter as a factor.

For the solution of the problem Bøggild's work is by far the most important recent contribution as he relates the phenomenon to chemical composition and definite crystal planes.

[16] *Kgl. Danske Videnskab. Selskab., Math.-Fys. Medd.*, VI, 3, 1924.

Summarizing the essential points that have been discovered in the past century we find:

(1) Iridescent color in crystals is reflected from definite crystallographical planes.
(2) In most cases the material that has been examined was twinned.
(3) The colors are interference colors usually of the first or second order.
(4) The color of the iridescent light is complementary to that of the transmitted light at a given angle of incidence.
(5) The colored light is not plane polarized.
(6) The atoms of crystals can be so arranged that light will be refracted in different ways.
(7) The iridescent colors in plagioclase are shown only by those of certain chemical composition.

In looking over the established points the writer has been impressed with the fact we have here all the essentials for a logical explanation of the phenomenon.

In approaching the solution of the problem it may be assumed (until disproved) that the ordinary laws of optics hold. If these laws do not apply then they are no longer general laws but special cases of more general laws. Three of these laws that apply are:

(1) The law of reflection:
The angle of incidence = the angle of reflection.
(2) The law of refraction (Snell's law):

$$\sin \text{ i}/\sin \text{ r} = n.$$

(3) The law for interference:

$$n\lambda = 2t\mu \cdot \cos \text{ i}.$$[17]

For purposes of comparison with more recent crystallographic work the last formula may be written:

$$n\lambda = 2t\mu \cdot \sin \theta,$$

[17] The laws just mentioned have been derived largely by testing isotropic material. At least two of them require a more general expression when applied to anisotropic media and there is disagreement as to the third. This has been taken into consideration in the discussion that follows. The really important point is that light, which has entered a plane parallel optical system, will on emerging follow a path which is parallel either with the incident ray or with the ray that is reflected from the first surface.

which is fundamental in the study of crystal structure by means of X-rays but as in this case the index of refraction is approximately unity, it has been modified by Bragg to the form:

$$n\lambda = 2d \cdot \sin \theta,$$

where d is the spacing between similar atomic planes. With monochromatic X-rays we get in this instance a monochromatic interference reflection. At other angles there is no reflection until suddenly we get a reflection of another order.

With glass and other amorphous substances the conditions for interference colors are met when:

$$n\lambda = 2t\mu \cdot \sin \theta.$$

The same formula will also apply to crystallized substances, but as:

$$t = md,$$

we can modify our formula so that it reads:

$$n\lambda = 2md\mu \cdot \sin \theta.$$

Of equal importance in approaching the solution is the formula which correlates the specific gravity, molecular weight, size of unit cell and the number of molecules in the unit cell

$$10^{-24} \times \text{S.G.} \times abc = \text{N(mol. wt.)} \times 1.65 \times 10^{-24}.$$

From our knowledge of the dimensions of the unit cells of crystals we can safely say that there is no possibility of interference reflections from single cells with ordinary light. A thickness such that we can obtain a retardation of a wave length of visible light is necessary, and in general it is by no means certain that an atomic plane that gives interference reflections with X-rays will give a reflection with ordinary light or that the reverse will hold.

The results obtained by numerous workers with X-rays show that we obtain interference reflections from a multitude of crystallographic planes which in general are placed at an angle to the visible reflecting surfaces. The work of von Hessel, Nordenskiöld, Brewster, Senff, the Rayleighs, and Bøggild has demonstrated that iridescent color is produced by reflection from definite crystallographic planes which in general make an angle with the apparent reflecting surface.

Inasmuch as we have interference reflections from definite crystallographic planes both with X-rays and with visible light we adopt the hypothesis that the laws governing both phe-

nomena are the same. We may then write the formula governing interference colors by reflection from crystals as:

$$n\lambda = 2md\mu \cdot \sin \theta.$$

The peristerite which is used for the present discussion was obtained from a feldspar quarry in Monteagle township, Hastings County, Ontario, where it is associated as a minor constituent with green microcline, flesh red microcline, quartz and biotite, and is of pegmatitic origin. It is twinned after the albite law so that a series of fine striations is visible on the basal cleavage. It also exhibits at times pericline twinning of a remarkable character so that it is readily seen in a hand specimen on the (010) cleavage. (Fig. 2).

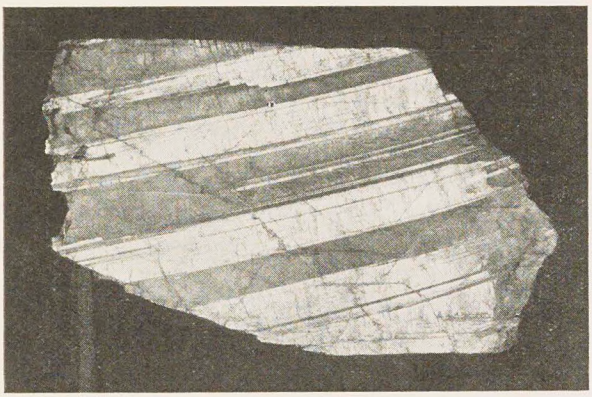

FIG. 2.

In the specimen that is illustrated the pericline lamellae have a maximum thickness of about half an inch.

The angle between the trace of the pericline twinning on (010) with the edge between (001) and (010) is 15°. The extinction in a plane perpendicular to (001) and (010) is 10°. Both these observations indicate a plagioclase with approximately 90 per cent albite.

In some of the specimens the central portion is iridescent while the outer border for a width, in some cases, of three fourths of an inch is non-iridescent. Occasionally there are iridescent portions in this outer zone. In view of Bøggild's statement that the peristerites contain about ninety per cent of the albite molecule, a sample of the iridescent portion of one of these specimens was carefully prepared and analyzed by M. C. Haller, research assistant

in the Department of Mineralogy, University of Toronto, with the following result:

	Per cent	Mol. pts.	Ab	Or	An	$H_2Al_2Si_2O_8$	Excess
SiO_2	66.25	1.104	.942	.096	.044	.002	
Al_2O_3	20.94	.205	.157	.016	.022	.010	
Fe_2O_3	0.34	.002					.002
CaO	1.23	.022			.022		
MgO	0.00	—					
Na_2O	9.74	.157	.157				
K_2O	1.50	.016		.016			
H_2O	0.20	.011				.011	
	100.20						
	Percentages		82.27	8.90	6.12	2.64	.34
S.G.	2.631						

This corresponds to 82.27 per cent of albite and 8.90 per cent of orthoclase or if we consider orthoclase as equivalent to albite we then have 91.17 per cent which is in accord with Bøggild's statement. The ratio of albite-orthoclase to anorthite is almost exactly eight to one and if we consider water to be equivalent to lime it is five to one. In either case we have a condition for a definite molecular arrangement.

For purposes of comparison another carefully prepared sample of the non-iridescent portion was analyzed by Mr. Haller and the results together with the recalculation follow:

	Per cent	Mol. pts.	Ab	Or	An	$H_2Al_2Si_2O_8$	Excess
SiO_2	66.72	1.111	.942	.072	.028	.018	.051
Al_2O_3	20.98	.206	.157	.012	.014	.009	.014
Fe_2O_3	0.31	.002					.002
CaO	0.81	.014			.014		
MgO	0.00	—					
Na_2O	9.74	.157	.157				
K_2O	1.12	.012		.012			
H_2O	0.16	.009				.009	
	99.84						
	Percentages		82.27	6.67	3.89	2.16	4.80
S.G.	2.628						

An interesting feature of this analysis is the percentage of albite which is exactly the same as in the iridescent material. The per-

centages of orthoclase, anorthite and $H_2Al_2Si_2O_8$ are all less. The striking difference, however, is in the excess of silica and alumina.

The ratio of albite-orthoclase to anorthite is 12 to 1 or, if we consider water as equivalent to lime, 8 to 1. The excess of silica and alumina, however, provides for a condition where a continuous regular molecular network would not be expected.

Measurement on the goniometer showed that a diffuse reflection was obtained from what is assumed to be an atomic plane which made an angle of $23°15' \pm 1°$ with the (010) face.

The specimen was then adjusted with a prismatic edge parallel to the axis of the vertical circle and the following measurements made:

	ϕ	ρ	
$(0\bar{1}0)$	180° 00′	90° 04′	
(010)	0° 02′	90° 04′	
$(\bar{1}\bar{1}0)$	118° 37′	90° 04′	
(110)	299° 12′	91° 00′	
$(00\bar{1})$	99° 43′	25° 21′	best base
(001)	81° 20′	25° 21′	
$(\bar{1}.15.\bar{3})$	14° 50′	71° 23′	iridescent reflection

The accuracy of the readings is not such that exact indices can be assigned as the reflection from $(\bar{1}.15.\bar{3})$ is diffuse and the position is susceptible to a variation of a degree or more in both the ϕ and ρ angles.

When the plane of the iridescent reflections was brought to the polar position there was no change in intensity in a complete rotation of the vertical circle.

The color in different parts varies but cannot be due to any known cleavage, for with incipient cleavage the Newton color rings are to be seen with ordinary conditions of incidence. The difference in color, however, appears to be due to differences in the thickness of the twinning lamellae. The color appears to be due principally to the first lamella, though some specimens show a fainter color when turned so as to look at the second lamella from the corresponding crystallographic direction.

The writer's conception, based on the three fundamental laws of optics mentioned above, of the path of the light causing the iridescent color is shown in figure 3.

The cause of the color may be summed up as follows:

The atoms (probably of calcium) are so spaced as a network or grating that the plane $(\bar{1}.15.\bar{3})$ (AB) acts as a reflecting and refracting plane for visible light, though it is extremely doubtful whether it would give a reflection with X-rays.

On entering the crystal the incident ray IA is refracted normally on AB not on MN and proceeds to the twinning plane where it is reflected normally from A″B″. On emerging from the crystal it is refracted by A′B′ and gives an interference color with I′A′ which is reflected at the plane A′B′ along the line A′R.

The particular tint of color is determined by the thickness of the twinning lamellae. This is shown more clearly on a polished surface where the crystal has been bent in places so that the thickness of the first lamella is not constant.

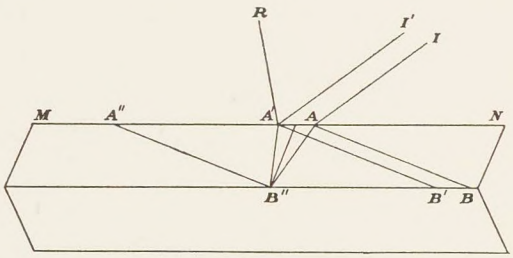

FIG. 3.

The common occurrence of twinning in the substances that exhibit iridescent color has naturally suggested that this plays an important part. Twinning provides for lamellae that are comparable in thickness with the wave lengths of visible light, and there is a discontinuity in optical arrangement at the twinning plane. Further than this the writer can see no reason for attributing the iridescent color to twinning.

In this paper an attempt has been made to give an explanation for iridescent color based upon well recognized laws. If the conclusions are incorrect there is a most attractive field to search for the fundamental laws of reflection, refraction and interference of light.

There are many points connected with iridescent color that remain to be solved. Many crystallized substances have been examined which exhibit this phenomenon. Where exact measurements have been made the color has been shown to be connected

with a definite crystallographic plane which in general makes an angle with the visible surface. In all cases it seems to be connected with a regular arrangement of atoms as was first suggested by Nordenskiöld.

The present paper is merely a beginning of the solution of the problems that are presented and it is premature to predict further developments but the field for study is extremely broad if we try to account for observed phenomena by physical laws.

If the laws of reflection, refraction and interference of light be correct, the work of numerous observers in the past century demonstrates most conclusively that visible light is refracted and reflected from atomic planes in crystals in a normal manner without reference to the surfaces.